

REMARKS

The instant Application was filed on January 23, 2004. In response to previous Office Actions, claims 21, 26 and 41-43 were canceled. Claims 1-20, 22-25, 27-40, 44 and 45 are at issue and have been rejected in the most recent Office Action. In view of the amendment to claim 44, the previously filed Terminal Disclaimer and the discussion hereinafter, Applicants submit that the present application is in condition for allowance and reexamination thereof is hereby requested.

The Office Action

The Office has rejected claims 1-20, 22-25, 27-40, 44 and 45 under 35 U.S.C. 102(e) as anticipated by or, in the alternative, under 35 U.S.C. 103(a) as obvious over U.S. Patent No. 6,699,457 ('457) or U.S. Patent No. 6,964,757 ('757) both to Cortright, et al.

The 35 U.S.C. 1-2(e)/103(a) Rejection

The Office has rejected claims 1-20, 22-25, 27-40, 44 and 45 under 35 U.S.C. 102(e) as anticipated by or, in the alternative, under 35 U.S.C. 103(a) as obvious over U.S. Patent No. 6,699,457 ('457) or U.S. Patent No. 6,964,757 ('757) both to Cortright, et al.

Cortright et al. teach a method of combining a catalytically enhanced thermal decomposition of an organic compound and a water gas shift reaction in a single reactor. Figure 6 of the Cortright reference (Note: All sites to Cortright made herein are to the '457 reference, but the same information is available in the '757 reference as well, just in a different location) depicts a diagram of this process.

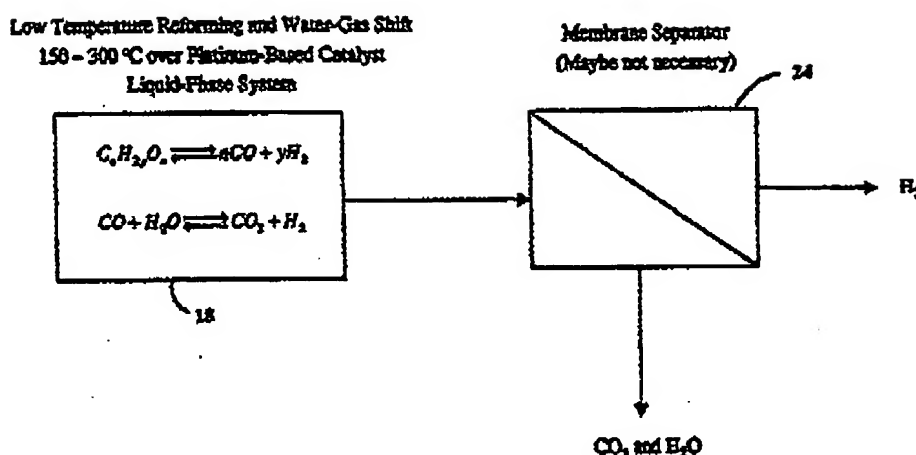


FIG.6

As can be seen, the organic compound $C_nH_{2y}O_n$ is converted to nCO and yH_2 in the initial reaction and thereafter the CO is reacted with H_2O to produce CO_2 and H_2 via a water gas shift (WGS) reaction. The products that finally come out of the reactor include H_2 gas, CO_2 gas, CO gas and excess water vapor (H_2O). Figure 6 shows the H_2 being separated from the rest of the gases using a membrane type separator. The invention of Cortright is that they were able to select certain organic compounds and certain catalysts such that the catalytic decomposition reaction used to convert the organic to CO and H_2 occurs at temperatures lower than the traditional steam reformation reaction. This is desirable because the WGS reaction works best at a lower temperature than the traditional steam reformation reaction. This is why the prior art processes (shown in figure 1 of Cortright) used separate reactors for the WGS reactions.

As can be seen from figure 6, the reactants for the production of hydrogen from the Cortright process are the organic material (which decomposes into CO and H_2) and H_2O

(which reacts with the CO that is formed to produce CO₂ and H₂). No other reactants are needed or called for by Cortright. Cortright uses a catalyst. Cortright also optionally uses a catalyst modifier. That is Cortright states (at column 14 lines 64 to 67) that , "The condensed liquid phase method of the present invention may also optionally be performed using a salt modifier that increases the activity and/or stability of the catalyst system" (emphasis added). It is important to note that the only part of the Cortright invention that uses any type of salt modifier is the "condensed liquid phase". Only Example 12 deals with the condensed liquid phase of the Cortright invention. Examples 1-5, 8 and 11 deal only with the production of suitable catalysts. Examples 6, 7, 9 and 10 relate to the gas phase reaction and as such do not include any "salt modifier" (i.e. no KOH). Therefore, as far as examples are concerned, only Example 12 relates to the use of a "salt modifier".

Cortright further states (at column 14, line 67 to Column 15, line 5) that the "salt modifier" for the catalyst is "Preferably . . . a water-soluble salt of an alkali or alkali earth metal" such as an ". . . alkali or an alkali earth metal hydroxide, carbonate, nitrate, or chloride salt." These alkali or alkaline earth catalyst modifiers are well known in the art. They have been known since at least 1969. See for example U.S. Patent No. 3,424,811 ('811). At column 3, lines 28-42 the '811 patent states:

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A critical feature of the catalyst compositions of the invention is the presence therein of a minor proportion of a compound of certain alkali metals as a catalyst modifier. Although the precise role of the alkali metal compound is not known with certainty, it is evident that the alkali metal plays a substantial role in determining the selectivity of the process. The preferred type of alkali metal compound to be employed is the oxide, although the alkali metal is suitably, and in fact is customarily provided in some other form of chemical combination, e.g., as the hydroxide, nitrate or the like, which is readily converted to the oxide. The choice of the alkali metal of the alkali metal compound is of some importance as compounds of all alkali metals are not equally effective in modifying the activity of the catalysts. The most suitable alkali metals are alkali metals of an atomic number from 19 to 55 inclusive, i.e., potassium, rubidium or cesium, although the lighter alkali metals sodium and lithium are also effective. Largely for reasons of economics, the use of compounds of potassium as the catalyst modifier is preferred.

Thus, it can be seen that the "salt modifier" for catalysts is well known in the art and clearly Cortright et al. knew what the salt modifier's role was in their process. As clearly stated, the salt modifier is to enhance the catalyst and does not comprise a reactant in their processes. Further evidence that the "salt modifier" (i.e. KOH) is not a reactant can be found at in Cortright at column 15, lines 5-8, where it states that "If an optional modifier is used, it should be present in an amount from about 0.5% to about 10% by weight as compared to the total weight of the catalyst system used" (emphasis added). Clearly the "salt modifier" is not a reactant, as its use is based on the weight of the catalyst and not on the weight of the organic reactant.

One very clear indicator that the optional "salt modifier" (KOH is used in example 12 in the specification) is merely acting as a catalytic enhancer can be deduced from Figure 16 of the Cortright references. Figure 16 shows that as the concentration of KOH is increased from zero up to 0.006 M, the production rate and selectivity of hydrogen production increase. However, when the concentration of KOH is further increased to

0.012 M, the production rate and selectivity do not further increase. (Once again, to be perfectly clear, this information can be deduced from Figure 16.) If KOH were actively reacting with $C_6H_{14}O_6$ (sorbitol) to form hydrogen, then the further increase in KOH concentration should have further increased both the production rate and the selectivity of hydrogen production.

Turning now to Example 12 of Cortright (the only example that deals with the condensed liquid phase reaction and as such the only example that includes a "salt modifier"), a 10 wt % aqueous sorbitol solution is reacted with the water in the solution, in the presence of 0.006 M KOH (the "salt modifier"). Analyzing the Office's contention that KOH is reacting with sorbitol to produce the hydrogen, we first make an assumption for the sake of calculation that 1000g of solution = 1 liter of solution and take a basis of 1000g solution.

Sorbitol

$C_6H_{14}O_6$ - 182.17g/mole

10 wt % sorbitol solution = 100g sorbitol /1000g solution.

Therefor given a basis of 1000 g solution, we have 100g or 0.549 mole sorbitol.

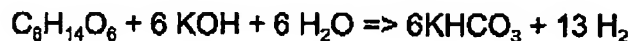
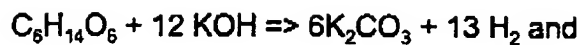
KOH

0.006 molar KOH solution = 0.006 mole/liter = 0.006 mole/1000g solution

Therefor given a basis of 1000 g solution, we have 0.006 mole KOH.

Thus, we have 549 mole sorbitol per 6 mole of KOH or 91.5 moles sorbitol per mole KOH.

The sorbitol reactions with KOH would be:



To make the carbonate byproduct, the stoichiometric ratio of sorbitol to KOH needs to be 1:12. Therefore to reach the proper stoichiometry, the 91.5 moles of sorbitol would need 1098 moles of KOH, in other words, the 0.549 moles/liter of Sorbitol would need 6.58 moles of KOH/liter. Thus to meet stoichiometry, the KOH would have to be 1100 times more concentrated than it is.

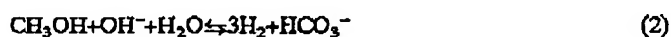
To make bi-carbonate byproduct, the stoichiometric ratio of sorbitol to KOH needs to be 1:6. Therefore to reach the proper stoichiometry, the 91.5 moles of sorbitol would need 549 moles of KOH, which means that the 0.549 moles/liter of Sorbitol would need 3.29 moles of KOH/liter. Thus to meet stoichiometry, the KOH would have to be 550 times more concentrated than it is.

The amount of KOH is clearly insignificant in comparison to the amount of sorbitol and the KOH does not contribute to production of hydrogen by the reaction of a base with an organic substance. As previously argued, and as is clearly stated in the Cortright references, the metal salt (KOH in the example) acts as a catalyst enhancer in the reactions thereof and does not directly contribute to the production of hydrogen by reaction with an organic substance.

The Office has suggested that the Cortright reference is not limited to example 12, but that there are many other examples and the specification as a whole must be addressed. As stated above, Example 12 is the only example that the salt modifier can be used. Further, the Office states that Cortright suggests that the pH of the Cortright reaction can be as high as 10.2 and the Office points to Cortright's statement that "It is much

preferred that the water and the oxygenated hydrocarbon are reacted at a pH of from about 4.0 to about 10.0." It is unclear how this translates to a pH of 10.2, but no matter, a pH of even 10.2 has an OH⁻ concentration of 0.00016 mol/liter. This is 37.5 times less hydroxide ion than Example 12 and would thus be even less likely to contribute to the production of hydrogen by reaction with an organic substance. Thus to meet stoichiometry, the KOH would have to be 20625 times more concentrated for the bicarbonate reaction and 41250 times more concentrated for the carbonate reaction.

The Office questions a statement in a previous patent of the instant inventors. The statement from U.S. Patent No. 6,607,707 states that:



controlling. Thermodynamic data shows that reaction (2) is controlling when the pH of the initial reaction mixture is between about 6.4 and 10.3 and that reaction (3) is controlling when the pH of the initial reaction mixture is above about 10.3. When the pH of the initial reaction mixture is below about 6.4, reaction (1) is controlling.

With respect to this statement, it should first be noted that:

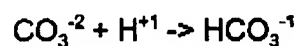
(see <http://en.wikipedia.org/wiki/Carbonate>)

In aqueous solution, carbonate, bicarbonate, carbon dioxide, and carbonic acid exist together in a dynamic equilibrium. In strongly basic conditions, the carbonate ion predominates, while in weakly basic conditions, the bicarbonate ion is prevalent. In more acid conditions, aqueous carbon

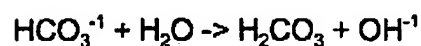
dioxide, $\text{CO}_2(\text{aq})$, is the main form, which, with water, H_2O , is in equilibrium with carbonic acid - the equilibrium lies strongly towards carbon dioxide.

Thus what the statement from the 6,607,707 is really talking about is this dynamic equilibrium. That is, at pH 10.3 and above, carbonate ions are stable, below pH 10.3 but above pH 6.4 bicarbonate ions are stable and below pH 6.4, carbonic acid is formed, which decomposes into CO_2 . Said in terms of equations:

Below pH 10.3 in aqueous solution, the carbonate ion converts into a bicarbonate ion.



Below pH 6.4 in aqueous solution, the bicarbonate ion converts into carbonic acid:

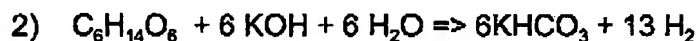
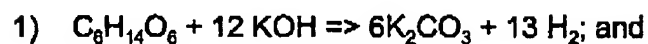


and decomposes: $\text{H}_2\text{CO}_3 \rightarrow \text{H}_2\text{O} + \text{CO}_2$

Thus, this information in 6,607,707 was based purely on the thermodynamics of the aqueous carbonate byproduct state. Further, it should be noted that it takes both thermodynamics and kinetics to make a chemical reaction occur. The thermodynamics discussed above only dictates which ending species is stable. It does not indicate that the reaction will actually proceed. That is further governed by kinetics. If one reactant species is not present in sufficient quantities, then the reaction will not proceed. For instance, in a mixture of hydrogen and oxygen, there is a specific threshold ratio under which the combustive reaction of hydrogen and oxygen will not proceed. The same is true for gasoline and air. This is because the kinetics for the reaction at those reactant concentration levels is not large enough. Therefore, reactions 2 and 3 will not proceed if the concentration of the reactants is too low, even though the thermodynamics may allow for stable end products of the reaction.

Further, even if we assumed (for the sake of argument) that all of the KOH did react with the Sorbitol in Example 12 of Cortright, and further assumed that the remaining Sorbitol reacted via the Cortright equations the amount of hydrogen produced by the KOH would be minuscule in comparison to the hydrogen produced by the real reaction of the Cortright process. The analysis is as follows:

The KOH reactions



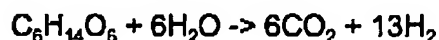
The ratio of sorbitol to KOH in Example 12 of Cortright, as calculated above is 549 mole sorbitol per 6 mole of KOH.

For equation 1), 6 mol KOH consumes 0.5 mol of sorbitol and makes 6.5 mol H_2 .

For equation 2), 6 mol of KOH consumes 1 mol of sorbitol and makes 13 mol H_2

The remaining sorbitol for equation 1) is 549 mol - 0.5 mol consumed by KOH = 548.5 mol sorbitol left and for equation 2) 549 mol - 1 mol consumed by KOH = 548 mol sorbitol left.

The Cortright Equations



Thus 548.5 moles sorbitol will make 7130.5 mol H_2 ; and 548 moles sorbitol will make 7124 mol H_2 . So:



Percent of H₂ produced by KOH

- 1) $6.5 \text{ mol H}_2 / 7137 \text{ Total mol H}_2 * 100 = 0.09\%$ (i.e. only 9 hundredths of a percent of the mol H₂ would have come from the KOH if it could have actually reacted), stated differently, 99.91% of the hydrogen would not have been produced by KOH reaction; and
- 2) $13 \text{ mol H}_2 / 7137 \text{ Total mol H}_2 * 100 = 0.18\%$ (i.e. only 18 hundredths of a percent of the H would have come from the KOH if it could have actually reacted), Thus 99.82% of the hydrogen would not have been produced by KOH reaction.

Realistically, the Cortright process could in no way be considered as "producing hydrogen by the reaction of a base with an organic material" because even if the reaction could proceed (which kinetics says it won't) it would not contribute in any meaningful way to the "production of hydrogen".

Further, regarding the Office's statement that there is no reason to believe that carbon dioxide would be produced in the Cortright process at a pH of 10.2, Applicant points once again to Example 12 of Cortright. The pH of a 0.006 molar KOH solution (as called for in Example 12) is 11.8. This is significantly higher than 10.2 and yet the offgas was 33% CO₂ (see column 20, lines 62-63). In fact even at an OH⁻ concentration of 0.012 molar which corresponds to a pH of 12.1, the production of CO₂ is not reduced (see Figure 16 of Cortright).

The Office has also suggested that the Cortright processes do not necessarily produce CO₂. In support of this, the Office points to Equations (6), (7), and (8) in column 8 of the '757 Cortright reference (which is also in column 8 of the '457 reference). However, the Office has misunderstood what Cortright was discussing in column 8. The reference to the equations pointed out by the Office states (at column 8 lines 12-18 of

'457):

"The thermodynamic trends shown in FIG. 2 also indicate that appropriate catalysts for use in the present invention must not show high activity for the cleavage of C—O bonds. Consider, for example, the steam reforming of ethanediol, reaction (6), followed by cleavage of the C—O bond in carbon monoxide to form methane and water, reaction (7), leading to the overall process given by reaction (8)"

Thus Cortright is discussing what would happen if the wrong catalyst was chosen and Cortright goes on to say (at column 8 lines 32-38 of '457):

"Therefore, a reforming catalyst that is readily able to cleave C—C and C—O bonds would convert ethanediol at low temperatures to a mixture of CO and CH₄, instead of the desired product mixture of CO and H₂. The CO and H₂ product mixture is preferred because it is followed by the production of CO₂ and H₂ by the WGS reaction." (Emphasis added).

Hence, Equations (6), (7), and (8) in column 8 of the Cortright references are undesired reactions and are to be avoided by the appropriate selection of catalyst. Also, Cortright again makes it clear that the final products of their overall inventive reaction are CO₂ and H₂.

Thus, with regard to claim 1, the Cortright references do not teach or suggest the production of hydrogen by reaction of the organic with a base, and the production of a carbonate and/or a bicarbonate by-product.

Turning next to amended claim 44, another aspect of the novel process of the present invention is claimed. That is, claim 44 is to a process for producing hydrogen gas

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by reacting an organic substance with a base (and optionally water) to form the hydrogen gas without any substantial production of carbon dioxide or carbon monoxide. This is a feature that is clearly not taught or suggested by Cortright. Cortright makes large quantities of carbon dioxide and/or carbon monoxide. This cannot be avoided in Cortright because the organic compound is converted to CO and H₂ in the initial reaction and thereafter the CO is reacted with H₂O to produce CO₂ and H₂ via a water gas shift (WGS) reaction. Thus, claims 44 and 45 are allowable over the Cortright references.

Applicants contend that Cortright neither teaches nor fairly suggests the hydrogen production processes of the instant claims and that the rejections under 35 U.S.C. 102(e)/103(a) are untenable. Applicants earnestly request withdrawal thereof.

Conclusion

Claims 1-20, 22-25, 27-40 and 44-45 remain at issue. In view of the amendments to the claims, the previously filed terminal disclaimer and the discussion herein above, Applicants submit the present application is now in condition for allowance. Applicants respectfully request timely notice of allowance thereof.

Should the Examiner have any comments or suggestions which would place the instant application in better condition for allowance, Applicants earnestly request the Examiner to contact the undersigned.

Date: March 18, 2008

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